GDP-0393

NDA93-106

NONDESTRUCTIVE ASSAY MEASUREMENTS OF ^{237}Np IN BUILDING K-33

SUMMARY REPORT

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September 9, 1993

Prepared by the
NDA Department
East Tennessee Technology Park
Oak Ridge, Tennessee 37831-7319
managed by
LOCKHEED MARTIN ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under Contract DE-AC05-84OR21400

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SUMMARY REPORT

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INTRODUCTION

In support of the K-25 Site Facilities Operations, a nondestructive assay (NDA) survey was conducted in Building K-33 at the Oak Ridge K-25 Site to locate and quantify residual ^{237}Np deposits which may present a transuranic handling concern. The NDA Department of the K-25 Site Technical Division performed the ^{237}Np measurements during April, May, and June 1993.

Neptunium was introduced into the enrichment cascade along with uranium in the form of reactor returns. Reactor returns are the uranium recovered by chemical processing of spent reactor fuel. The neptunium created in the reactor fuel is not completely removed in the processing of the spent fuel and some will remain in the material fed back into the cascade. Smith[1] and Bailey[2] have written reports discussing the history of reactor returns feed at the Paducah Gaseous Diffusion Plant (PGDP) and Oak Ridge Gaseous Diffusion Plant (ORGDP). Details in the two reports differ, but in general it can be said that reactor returns were fed to the ORGDP (now the K-25 Site) at various times from the 1950s through the early 1980s. Virtually all of the returns fed before 1970 were from plutonium production reactors at Hanford and Savannah River. After 1969, reactor returns from France and England were also fed into the cascade.

Potentially both neptunium and plutonium were introduced into the cascade in the reactor returns. Uranium, neptunium, and plutonium all form hexafluoride compounds, but the stability of the compounds differs greatly. Upon contact with the metal walls of the equipment, each of the compounds is reduced to tetrafluoride or other compounds which are not volatile at cascade temperatures and pressures. Uranium hexafluoride is more stable than either plutonium or neptunium hexafluoride. Plutonium hexafluoride is most readily reduced to plutonium tetrafluoride by contact with metal walls and rarely got farther into the cascade than the initial feed piping. Neptunium hexafluoride is less readily reduced. While concentrated in the immediate area of the feed points, neptunium spread throughout the cascade.

Bailey[2] calculated that 2.2 kg ^{237}Np and 44 g ^{239}Pu were received at the ORGDP in the form of reactor returns. A large fraction of the neptunium and plutonium remained in the feed cylinders and was not fed into the cascade. The quantities fed into the cascade are estimated to be 540 g ^{237}Np and 65 mg ^{239}Pu. Work performed at PGDP[1] and ORGDP[2] showed that neptunium hexafluoride readily reacted with process equipment surfaces and uranium compounds in the system and, when reduced, formed compounds that are not volatile in the range of cascade operating temperatures. Those compounds were found to be present as dust on the surfaces of equipment removed from the cascade. These dust particles did not pass through the barrier tubes, but were transported in the B process stream and tended to collect or settle out in the stagnant regions. The neptunium hexafluoride that had not been reduced gradually worked its way throughout the cascade. Sampling[1] of product and tails cylinders at the PGDP has found only trace amounts of ^{237}Np in the product cylinders and no detectable quantity of ^{237}Np in tails cylinders.

Building K-33 contains eight units with ten cells in each unit. Each cell consists of eight stages containing a size "000" converter and compressor. Bailey[2] assumed that nearly all of the neptunium that was fed to the cascade was removed when the compressors and converters were exchanged in the cascade

improvement programs. Measurements show that portions of the neptunium remain in the cascade piping and equipment, primarily in the B-line vacuum balancers and the reactor returns feed lines. The NDA survey indicates that nearly all of the compressor B-line vacuum balancers contain deposits of neptunium. The deposits are probably composed of particulate matter that was swept into the vacuum balancers through the connection to the curved section of the B-line pipe between the control valve and the compressor. The B-line pipe connects the B outlet on the converter to the compressor of the previous stage. The deposits in the reactor returns feed lines are a result of the reduction of the neptunium hexafluoride through contact with the pipe walls.

MEASUREMENT APPROACH

The measurement survey consisted of two major components: (1) isotopic mapping to identify locations where neptunium was present to establish the extent of the spread of the neptunium throughout the building and to establish the ratio of ^{237}Np to uranium at each location and (2) quantitative measurement of the uranium contained in the deposits with the highest concentration of ^{237}Np. The quantity of ^{237}Np in each of the quantitatively measured deposits was calculated from the measured quantity of uranium and the ^{237}Np-to-uranium ratio. A previously reported survey of Building K-33[3] displayed the distribution of detectable ^{237}Np in the pipe gallery. Figure 1 illustrates the results of that survey.

Isotopic mapping involves collecting gamma-ray spectra using high-resolution gamma-ray measurement equipment and analyzing the spectra to determine: (1) the ^{235}U concentration in the uranium contained in the measured process equipment, (2) the ratio of the mass of ^{237}Np to the mass of uranium if neptunium is present, and (3) the presence of other isotopes which can interfere with quantitative NDA measurements (e.g., ^{232}U). The results of the analysis of the spectra collected throughout the building were used to map the distribution of ^{237}Np in the building. The quantitative measurements were also made using high-resolution gamma-ray measurement equipment. While spectra collected for the purpose of isotopic mapping are collected as close as possible to the measured item to minimize the data collection time, quantitative measurements must be made with the detector farther away from the measured item to reduce geometrical effects of the distribution of the deposit in the item. More efficient low-resolution equipment could not be used because the gamma rays from the ^{237}Np and ^{232}U in the deposits interfere with the ^{235}U measurement.

RESULTS

High-resolution gamma-ray spectra were collected at 266 locations for isotopic mapping purposes. Spectra taken early in the program indicated that the B-line vacuum balancers were likely places to find ^{237}Np concentrations. The vacuum balancers have relatively thin walls and are uniform throughout the whole cascade except for Stage 8 in each cell. Therefore, measurement of the equivalent vacuum balancer in each cell gave a picture of the distribution of the ^{237}Np in the cascade. As shown in Fig. 2, the vacuum balancers are cylindrical with a diameter of 31 in. and a height of 12 in. The convolutions which allow for expansion are covered with a steel boot. The diameter including the boot is 38 in. Figure 3 shows the results in parts per million ^{237}Np relative to the total uranium from the spectra of the Stage 1 vacuum balancer in all 80 cells in the building. In addition, spectra were collected from the vacuum balancers in all the stages of Cells K-902-6-6 and K-902-6-10. The higher ^{237}Np concentrations in these cells implies that much of the reactor returns were fed to cells in Unit K-902-6. Evidence of ^{237}Np was found in the spectra collected in the reactor returns feed vaporization room and in the feed lines coming from the feed

room. Some sections of the feed headers in the reactor returns feed vaporization room and the pipe galleries were not accessible for isotopic mapping. Of the 266 spectra, 160 showed detectable quantities of ^{237}Np.

According to R. R. Redmond of the Health Physics Department, transuranic release limit for surfaces must be followed for deposits if the ratio of the disintegration rate of the uranium to the disintegration rate of the transuranic component (e.g., ^{237}Np) of the deposit is less than 50. For airborne material, the transuranic release limit is a uranium-to-transuranic disintegration ratio of less than 300. The isotopic mapping of Building K-33 found 13 deposits with a ratio of less than 50 and 55 deposits with a ratio of less than 300. Most of the deposits with significant uranium-to-^{237}Np disintegration ratios were either reactor returns feed lines or B-line vacuum balancers. All of the deposits with ratios less than 85 were quantitatively measured with the exception of one deposit in the area of the Unit 3 to Unit 6 crossover. Four vacuum balancers and an evacuation line crossover with ratios between 85 and 300 were measured, and three vacuum balancers with large uranium deposits but ratios higher than 300 were measured. While time did not allow for measurement of all items with ratios less than 300, as many as possible were measured in the time available.

Table 1 summarizes the results of the quantitative measurements on 14 B-line vacuum balancers. Estimated quantities are given in grams for the uranium and ^{235}U and milligrams for ^{237}Np. Table 2 summarizes the results of the quantitative measurements on the feed headers and other pipes. Figures 4 and 5 illustrate the location of the quantitatively measured deposits in the A and B feed headers. (The figures are not to scale.) At one location, the A and C feed headers were too close together to be measured separately, but most of the deposit appeared from scanning data to be in the A feed header. An estimated total of about 750 mg ^{237}Np was found in all of the deposits measured. In addition, traces of ^{237}Np were indicated in two compressors and six converters in Units K-902-4, K-902-6, and K-902-8. The uranium-to-^{237}Np disintegration ratio in the compressor in K-902-6-10-7 is estimated to be 122. Time did not permit a complete survey of all compressors and converters. Indications of ^{237}Np were found in both coolers for the evacuation booster station. Extrapolating over the entire building gives an estimated maximum of between 1 and 2 g ^{237}Np remaining in Building K-33.

No indication of plutonium has been found in any of the 266 spectra collected in Building K-33.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions drawn from the isotopic mapping are as follows:

1. Concentrations of ^{237}Np are greatest in the feed lines both in the feed vaporization room and in the pipe gallery on the cell floor above the feed vaporization room.
2. Within the cascade, ^{237}Np concentrations are highest near the feed points, with deposits of ^{237}Np present in all of the 640 stages in Building K-33.
3. The compressor B-line vacuum balancers contain the highest concentrations of ^{237}Np in the building outside the feed lines, but the mass of the deposits is small.
4. ^{237}Np is also present in the evacuation booster stations and some of the evacuation piping.
5. No ^{237}Np was detected in the seal exhaust piping, the chemical trap area, or the cold recovery area.
6. Measurements of the two cobalt difluoride traps and the piping in the trap preheater room indicated the presence of uranium but did not detect ^{237}Np, although deposits of ^{237}Np were found in the feed line piping immediately downstream from the traps. Therefore, the cobalt difluoride trapping facilities

in the reactor returns feed vaporization room apparently were ineffective in removing ^{237}Np from the feed stream.

The quantitative results support the assumption that most of the ^{237}Np was removed from the cascade when converters and compressors were replaced during the cascade improvement programs. Although the quantities are small, high concentrations of ^{237}Np still remain in the reactor returns feed lines in the feed vaporization room and in the feed line piping going to the cell floor. The B feed line leaving the feed vaporization room contains more than 80% of the ^{237}Np found in this survey. Milligram quantities of ^{237}Np are found in the B-line vacuum balancers in almost all of the 640 stages in Building K-33. Previous experience[1] indicates that small quantities of ^{237}Np below measurable limits can be expected as a thin layer of dust spread on the inner surfaces of piping and equipment throughout the entire building. Since the ^{237}Np was detected throughout the entire K-33 building, it is reasonable to assume that some ^{237}Np may have moved to Building K-31. Some measurements should be performed in K-31 to determine if ^{237}Np is present in large enough concentration to require a complete survey of the building. Buildings K-131 and K-1131, which were used as feed buildings, and Building K-1410, which at one time was used to convert reactor returns oxide to hexafluoride, should be surveyed for the presence of ^{237}Np. If ^{237}Np is found in any of these buildings, the tie lines from these buildings to the cascade should also be surveyed. All UF_6 cylinders which were used for transfer of reactor returns have the potential for large concentrations of ^{237}Np, because as much as 75% of the ^{237}Np of the reactor returns was not transferred to the cascade. Building K-1420 is currently being surveyed by the NDA Department. Preliminary data indicate that the cylinder wash station in K-1420 has a significant concentration of ^{237}Np. ^{237}Np was found in the autoclave area of Building K-1423 when it was surveyed in 1991.[4]

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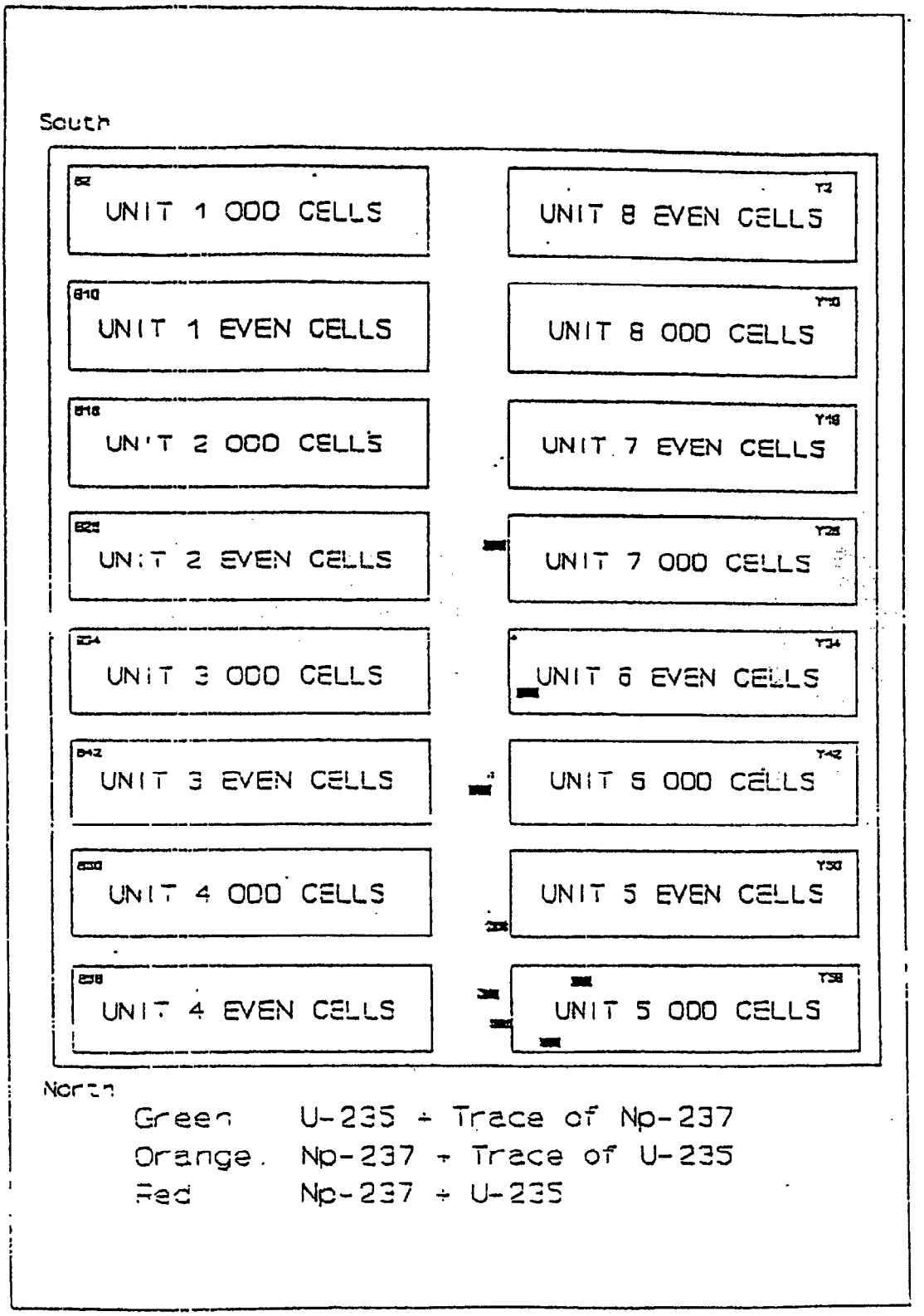

Fig 1 Location of ^{237}Np Deposits in the Building K-33 Pipe Gallery

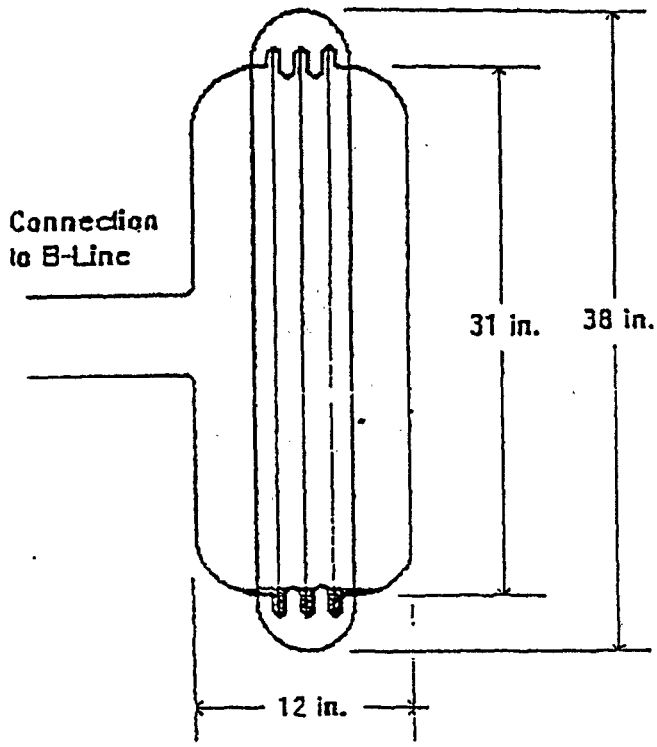

Fig. 2. Diagram of Typical B-Line Vacuum Balancer

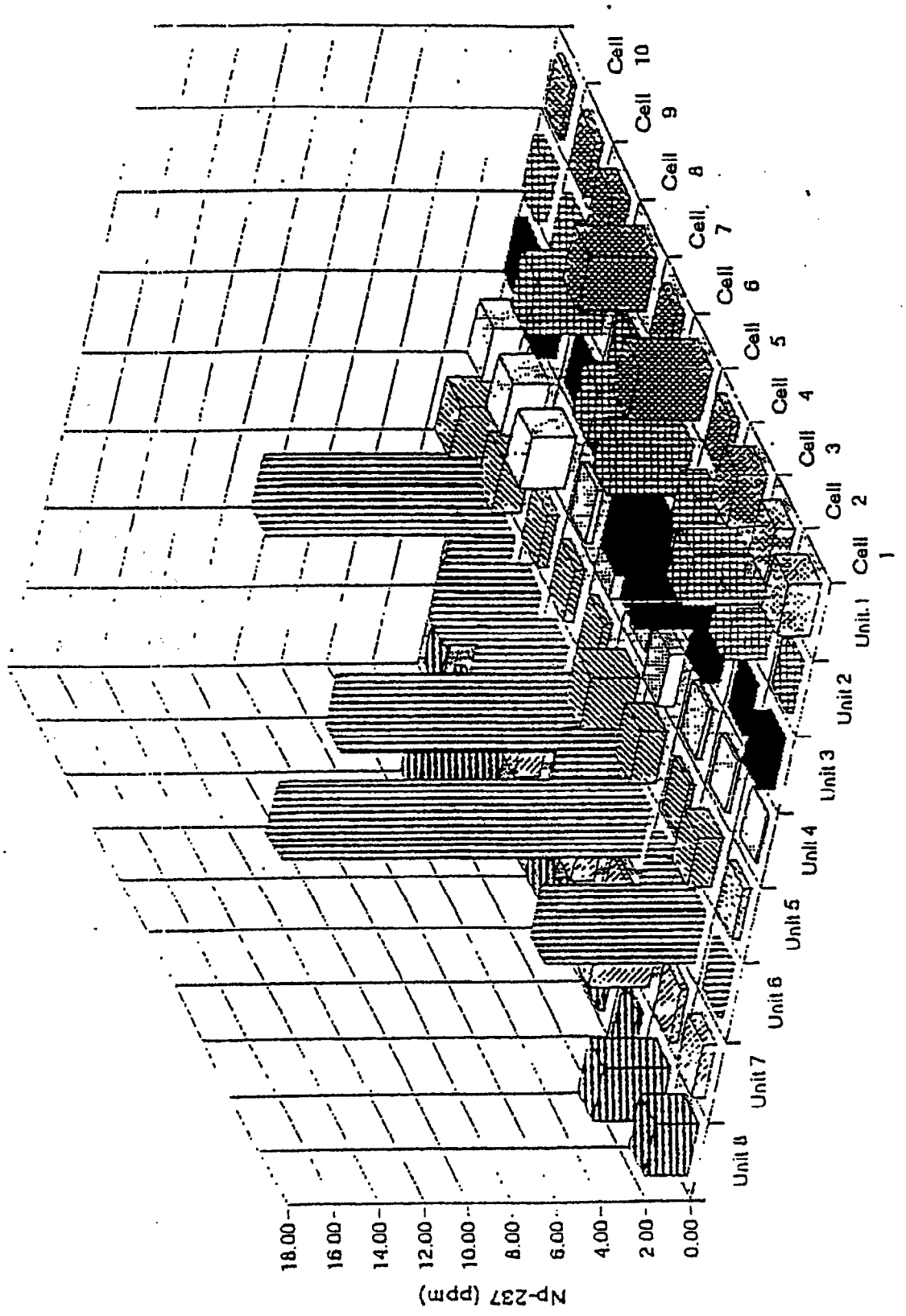

Fig. 3 Concentration of ^{237}Np in Stage 1 B-Line Vacuum Balancers

Table 1. Results of NDA ^{237}Np Survey of B-Line Vacuum Balancers

Location	U:^{237}Np (dps ratio)	^{235}U (g)	Uranium (g)	^{237}Np (mg)
K-902-1-9-1	1,347.8	5.3	913	0.3
K-902-2-8-1	143.8	2.1	388	1.3
K-902-2-9-1	1,197.4	6.8	1,224	0.5
K-902-5-1-1	1,426.4	8.8	1,018	0.4
K-902-6-2-1	75.9	0.7	75	0.5
K-902-6-4-1	31.8	0.2	20	0.3
K-902-6-6-1	46.1	0.5	58	0.6
K-902-6-6-3	67.4	1.3	136	1.0
K-902-6-7-1	157.6	1.2	138	0.4
K-902-6-8-1	129.6	1.5	159	0.6
K-902-6-10-1	53.8	0.3	60	0.6
K-902-6-10-3	82.8	0.9	101	0.6
K-902-6-10-5	88.3	0.6	71	0.4
K-902-6-10-7	11.8	0.1	13	0.6
Total	-	30.3	4,374	8.1

Table 2. Results of NDA ^{237}Np Survey of Piping in Building K-33

Location	Description	Diameter (in.)	Length (in.)	Figure Number[a]	Location of Deposit	U:^{237}Np (dps ratio)	^{235}U (g)	Uranium (g)	^{237}Np (mg)
Vertical Duct from Feed Room to Pipe Gallery	A and C Feed Header	4	48	4	1	11.9	7.9	888	39
	B Feed Header (Top)	4	48	4	2	0.7	0.7	117	81
	B Feed Header (Bottom)	4	96	4	3	0.6	2.1	328	286
Pipe Gallery near Crossover between Units 4 and 5	A Feed Header	4	32	5	4	31.5	9.1	1,251	20
		4	48	5	5	31.5	3.1	517	8
		4	9	5	6	13.8	1.7	392	14
	B Feed Header	4	36	5	7	4.1	2.2	217	27
		4	9	5	8	2.7	4.2	547	104
		4	74	5	9	4.1	4.9	436	53
		4	9	5	10	2.7	1.4	347	66
	Reactor Returns Tie Line	4	48	-	-	62.3	2.6	341	3
	Evacuation Line Crossover	12	326	-	-	210.2	56.5	7,528	18
Reactor Returns Feed Vaporization Room	Lines in Lower Duct	4	48	-	-	5.6	1.4	180	16
	Lines Leaving CoF_3 Trans	4	48	-	-	63.8	3.7	379	3
Total		-	-	-	-	-	101.5	13,468	738

[a] Refer to this figure for the location of the measured deposit.

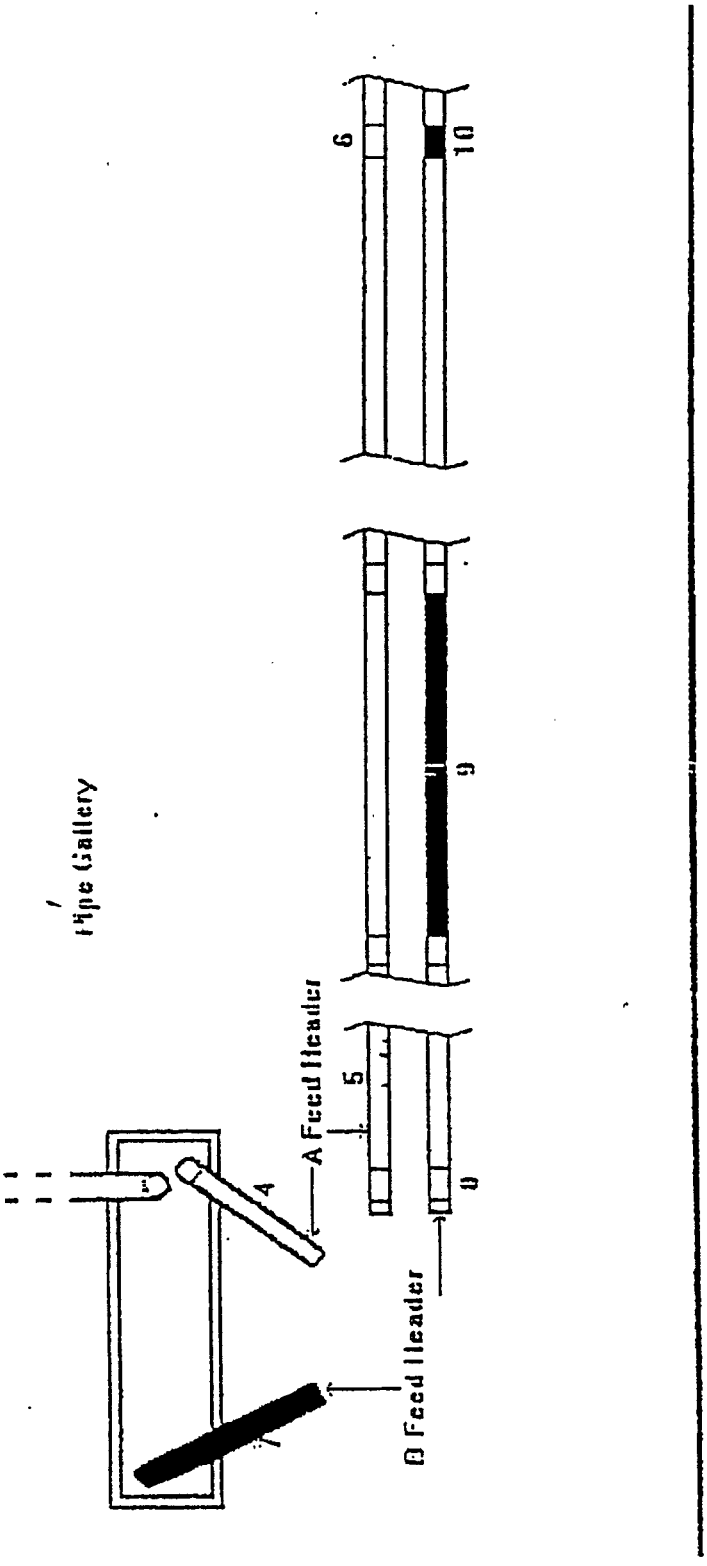

Fig. 5 Location of Measured Deposits in the Pipe Gallery